Supplemental Figure Captions

Supplemental Table S1. [a] K_a (M^{-1}) values reported are the mean values from at least three DNase I footprint titration experiments. Assays were performed at 22°C in a buffer of 10 mM Tris-HCl, 10 mM KCl, 10 mM $MgCl_2$, and 5 mM $CaCl_2$ at pH 7.0. Imidazole and pyrrole are shown as filled and non-filled circles, respectively; β-alanine is shown as a diamond; the dimethylaminopropylamide tail is shown as a half-circle with a plus; the achiral γ-aminobutyric acid turn residue is shown as a semicircle connecting the two subunits; and the chiral diaminobutyric acid turn residue is shown as a semicircle linked to a half-circle with a plus connecting the two subunits.

[b] This paper, Table 1

[c] This paper, Supplemental Figure S3

[d] Previously unpublished data

Supplemental Figure S1. Plasmid design for pCFH2, pCFH3, pCFH4, pCFH5, pPh2, and pMFST, indicating the designed match and mismatch sites for hairpin polyamides **12**, **13**, **14**, **16**, **17**, **19**, **21**, **24**, and **27**. Imidazole and pyrrole are shown as filled and non-filled circles, respectively; β-alanine is shown as a diamond; the dimethylaminopropylamide tail is shown as a half-circle with a plus; and the chiral diaminobutyric acid turn residue is shown as a semicircle linked to a half-circle with a plus connecting the two subunits.

Supplemental Figure S2. Quantitative DNase I footprint titration experiments for polyamides **13**, **14**, **16**, **17**, **19**, **21**, **24**, and **27** on the 295 bp, 5'-^{32}P-end-labeled PCR

product of plasmids pCFH2, pCFH3, pCFH4, pCFH5, and pPh2: lane 1, intact DNA; lane 2, G reaction; lane 3, A reaction; lane 4, DNase standard; lanes 5-15, 10 pM, 30 pM, 100 pM, 300 pM, 1 nM, 3 nM, 10 nM, 30 nM, 100 nM, 300 nM, 1 μM, respectively. Binding isotherms for the four designed sites are shown below each footprinting gel; θ_{norm} values were calculated according to published methods.[1] A binding model for the hairpin motif is shown above each gel with the polyamide bound to its target DNA sequence. Imidazole and pyrrole are shown as filled and non-filled circles, respectively; β-alanine is shown as a diamond; the dimethylaminopropylamide tail is shown as a half-circle with a plus; and the chiral diaminobutyric acid turn residue is shown as a semicircle linked to a half-circle with a plus connecting the two subunits.

Supplemental Figure S3. Quantitative DNase I footprint titration experiments for polyamides **6**, **12**, **20**, **22**, and **23**. A binding model for the hairpin motif is shown above each gel with the polyamide bound to its target DNA sequence. Imidazole and pyrrole are shown as filled and non-filled circles, respectively; β-alanine is shown as a diamond; the dimethylaminopropylamide tail is shown as a half-circle with a plus; and the chiral diaminobutyric acid turn residue is shown as a semicircle linked to a half-circle with a plus connecting the two subunits.

References

(1) Trauger, J. W.; Dervan, P. B., *Methods Enzymol.* **2001,** 340, 450-466.